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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

JUN KAMATANI ET AL.

Application No.: 10/073,012

Filed: February 12, 2002

For: LUMINESCENCE DEVICE AND DISPLAY APPARATUS

Examiner: M. Yamnitzky

Group Art Unit: 1774

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER TITLE 37 C.F.R. § 1.132

I, JUN KAMTANI, declare that:

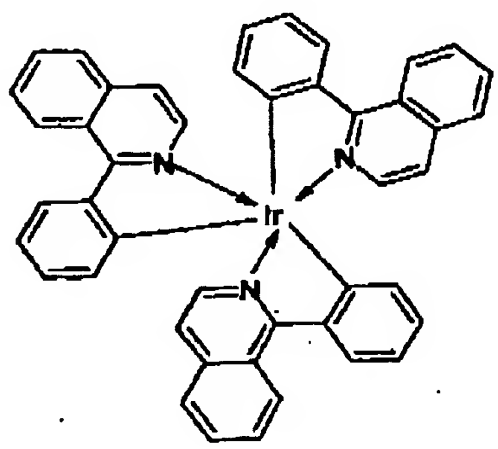
1. I reside at 17-20-202, sakuragaoka 5-chome, setagaya-ku Tokyo, Japan.

2. I have been a Research Scientist at Canon Kabushiki Kaisha since 1999.

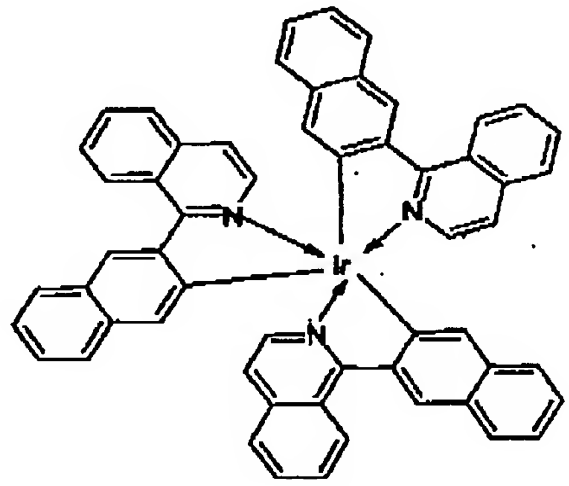
3. I have received an undergraduate degree in Department of Bioengineering from Tokyo Institute of Technology University in 1996, a graduate degree in Research Laboratory of Resources Utilization from Tokyo Institute of Technology University in 1999.

4. I am an inventor of the subject patent application and am familiar with the prosecution history of the subject patent application.

5. I have conducted tests to illustrate that the properties of Example Compound No. 1 in the above-identified application:



are superior to those of Example Compound No. (1-60) in U.S. Application Publication No. 2001/0019782 A1 (Igarashi):

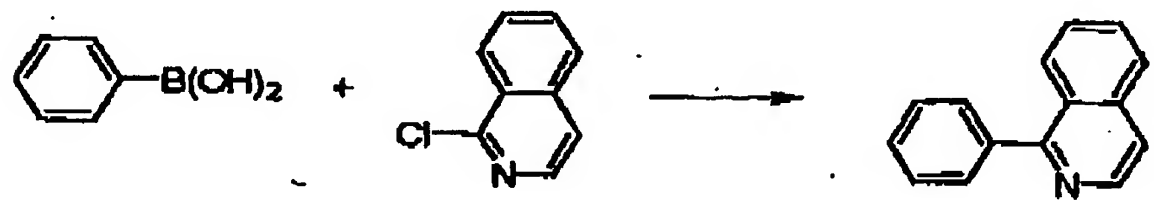


6. Preparation of the compounds

(A) Preparation of Example Compound No. 1 of the present invention

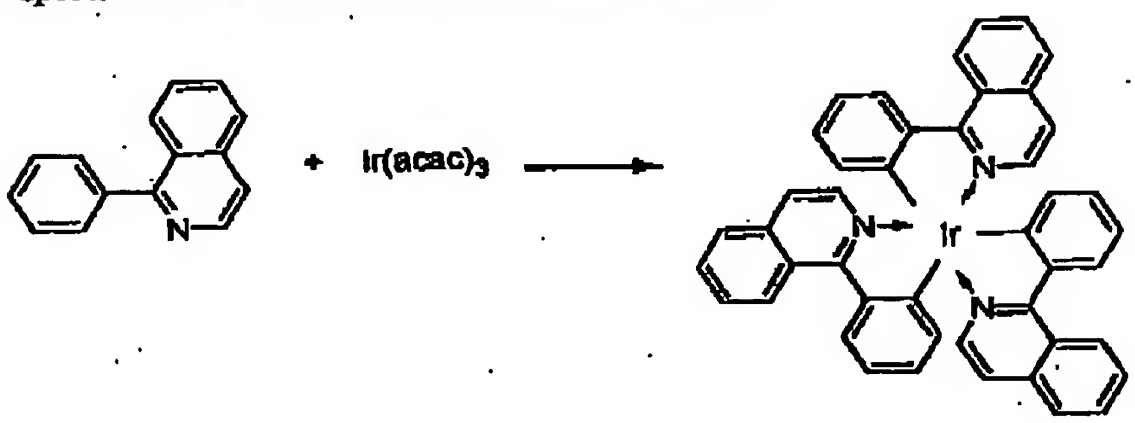
Example Compound No. 1 was prepared in accordance with the procedures outlined in Example 7 of the present application.

69.3 g (448 mmol) of isoquinoline N-oxide (made by Tokyo Kasei) and 225 ml of chloroform were placed and dissolved in a 1 liter-three-necked flask, and under stirring and cooling with ice, 219.6 g (1432 mmol) of phosphorus oxychloride was gradually added drop-wise thereto while the internal temperature was held at 15 - 20 °C. Thereafter, the temperature was raised, and reflux under stirring was performed for 3 hours. The reaction product was cooled by standing to room temperature and poured into iced water. After extraction with ethyl acetate, the organic layer was washed with water until neutrality, and the solvent was removed under a reduced pressure to provide a dry solid, which was then purified by silica gel column chromatography (eluent: chloroform/hexane = 5/1) to obtain 35.5 g (yield: 44.9 %) of 1-chloroisoquinoline white crystal.



In a 100 ml-three-necked flask, 3.04 g (24.9 mmol) of phenylboronic acid (made by Tokyo Kasei), 4.0 g of (25.0 mmol) of 1-chloroisoquinoline, 25 ml of toluene, 12.5 ml of ethanol and 25 ml of 2M-sodium carbonate aqueous solution were placed and stirred at room temperature under a nitrogen stream, and 0.98 g (0.85 mmol) of tetrakis (triphenyl-phosphine)palladium (0) was added thereto. Thereafter, reflux under stirring was performed for 8 hours under the nitrogen stream. After completion of the reaction, the reaction product was cooled and extracted by addition of cold water and toluene. The organic layer was washed with saline water and dried on magnesium sulfate, followed by removal of the solvent under a reduced pressure to provide a dry solid. The residue was purified by silica gel column chromatography (eluent: chloroform/methanol = 10/1) to

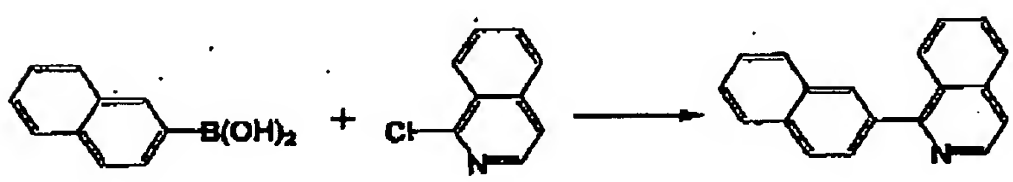
obtain 2.20 g (yield = 43.0 %) of 1-phenylisoquinoline. Figure 7 shows a ^{1}H-NMR spectrum of a solution of the compound in heavy chloroform.



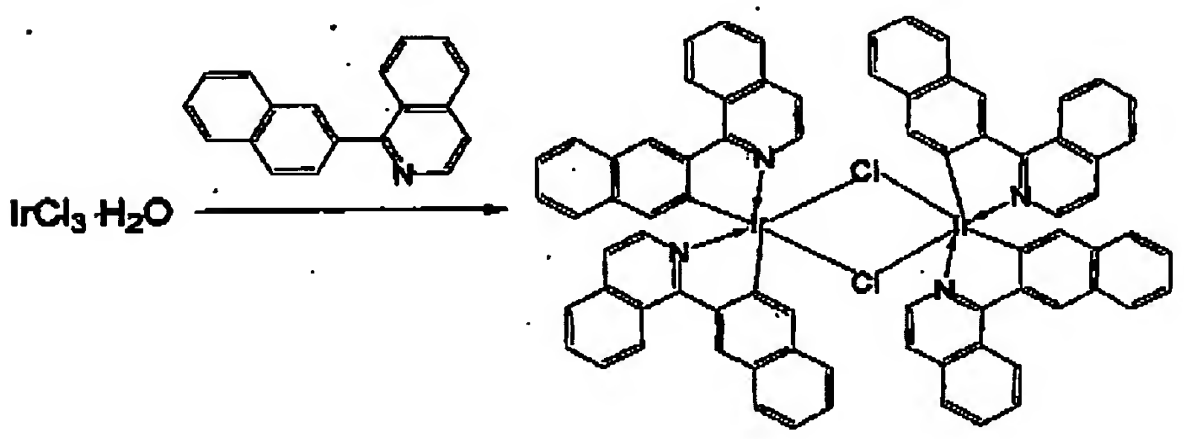
In a 100 ml-four-necked flask, 50 ml of glycerol was placed and heated at 130 - 140 °C under stirring and bubbling with nitrogen for 2 hours. Then, the glycerol was cooled by standing down to 100 °C, and 1.03 g (5.02 mmol) of 1-phenylisoquinoline and 0.50 g (1.02 mmol) of iridium (III) acetylacetonate (made by Strem Chemicals, Inc.) were added, followed by 7 hours of heating around ±210 °C under stirring and a nitrogen stream. The reaction product was cooled to room temperature and injected into 300 ml of 1N-hydrochloric acid to form a precipitate, which was filtered out and washed with water. The precipitate was purified by silica gel column chromatography with chloroform as the eluent to obtain 0.22 g (yield = 26.8 %) of red powdery tris (1-phenylisoquinoline-C^2,N)iridium (III) (Example Compound No. 1 of invention).

(2) Preparation of Example Compound No. (1-60) of Igarashi

Example Compound No. (1-60) (1-(2-naphtyl isoquinoline) was prepared in the following manner.

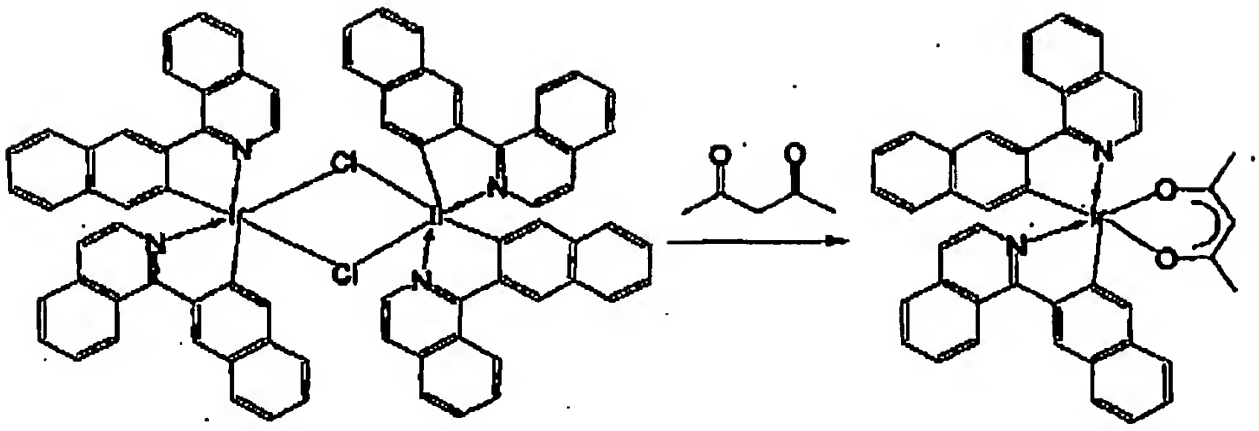


In a 200 ml-three-necked flask, 5.16 g (30.0 mmol) of 2-naphthyl-boronic acid (made by Aldrich Co.), 4.91 g (30.0 mmol) of 1-chloroisoquinoline, 50 ml of toluene, 25 ml of ethanol, and 50 mol of 2M-sodium carbonate aqueous solution were placed and stirred at room temperature under nitrogen stream, and 1.15 g (1.0 mmol) of tetrakis (triphenyl-phosphine)-palladium (0) was added thereto. Thereafter, reflux under stirring was performed for 8 hours under a nitrogen stream. After completion of the reaction, the reaction product was cooled to precipitate a crystal. The crystal was recovered by filtration and washed with water, followed by purification by silica gel column chromatography (eluent: chloroform/hexane = 3/1) and recrystallization from ethanol to obtain 5.21 g (yield = 68.0 %) of 1-(2-naphtyl)isoquinoline.

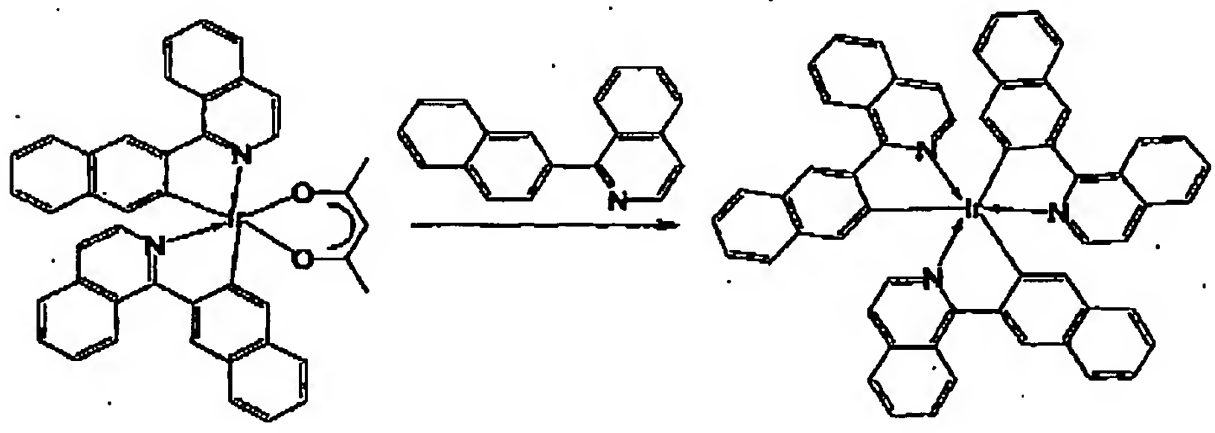


In a 300 ml-three-necked flask, 1.89 g (4.99 mmol) of trichloroiridium (III) hydrate, 2.81 g (11.0 mmol) of 1-(2-naphtyl)isoquinoline, 90 ml of ethanol, and 30 ml of water were placed and stirred for 30 minutes at room temperature under a nitrogen stream, followed by 8 hours of reflux under stirring. The reaction product was cooled to room temperature, and 20 ml of water was added. The precipitate was recovered by filtration and washed with water, followed successive washing with ethanol and acetone. After drying under a reduced pressure at room temperature, 2.48 g (yield = 78.1 %) of dark red powdery tetrakis[1-(2-naphtyl) isoquinoline-C^3,N]-(μ-dichloro)diiridium (III)

was obtained.



In a 200 ml-three-necked flask, 100 ml of ethoxyethanol, 2.20 g (1.49 mmol) of tetrakis[1-(2-naphtyl) isoquinoline- C^3,N]-(μ-dichloro)diiridium (III), 0.45 g (4.49 mmol) of acetylacetone and 2.20 g (2.06 mmol) of sodium carbonate were placed and stirred for 1 hour at room temperature under a nitrogen stream and then refluxed under stirring for 15 hours. The reaction product was left standing to cool, and 50 ml of water was added. The precipitate was recovered by filtration and washed with water. The precipitate was then washed with ethanol and dried under reduced pressure to obtain 1.70 g (yield = 70.9 %) of dark red powdery bis[1-(2-naphtyl)isoquinoline- C^3,N](acetylacetonato)-iridium (III).



In a 100 ml-three-necked flask, 0.64 g (2.51 mmol) of 1-(2-naphtyl)isoquinoline, 0.80 g (1.00 mmol) of bis[1-(2-naphtyl)isoquinoline-C^3,N](acetylacetonato)-iridium (III), and 50 ml of glycerol were placed and heated to

about 180 °C under stirring and a nitrogen stream. The reaction product was cooled to room temperature and poured into 120 ml of 1N-hydrochloric acid, and the precipitate was recovered by filtration and washed with water. The precipitate was purified by silica gel column chromatography with chloroform as the eluent to obtain 0.44 g (yield = 46.1 %) of dark red powdery tris [1- (2-naphtyl)-isoquinoline-C^3,N]iridium (III) (Example Compound No. (1-60) of Igarashi).

7. Comparative Test A

With respect to each of Example Compound Nos. 1 and (1-60), a toluene solution was prepared so that it provided an absorbance of 0.05 at an excitation wavelength of 450 nm in the ultraviolet-visible adsorption spectrum. Each of 2-3 ml of these solutions was placed in a fluorescent cell, and bubbling with argon gas was performed for 30 minutes in order to remove dissolved oxygen causing extinction of phosphorescence of the complex to obtain a solution saturated with argon gas. With respect to each of the compounds, an emission spectrum at the excitation wavelength of 450 nm was measured by a fluorescent spectrophotometer to obtain an emission peak wavelength. Further, on the basis of a luminescence quantum yield of 0.40 for $Ir(ppy)_3$ (described in "J. Am. Chem. Soc.", Vol. 107, pages 1431 and 1432 (1985)), a luminescence quantum yield of each compound was calculated as a relative value of an emission spectrum area. Similarly, a spectral luminous efficiency at each of emission peak wavelengths of 619 nm and 646 nm was calculated proportionally on the basis of a value described in Noboru Ota, "Color Engineering", Vol. 2, page 12, published by Tokyo Denki University Press. The results are shown in Table 1.

Table 1

	Ex. Comp. No. 1 of the Invention	Ex. Comp. No. (1-60) of Igarashi
(a) Emission peak wavelength	619 nm	646 nm
(b) Luminescence quantum yield	0.24	0.09
(c) Spectral luminous efficiency	0.393	0.134

(a) An emission peak required for red of the three primary colors (red, green, and blue) of the NTSC color system used in an organic electroluminescence display is 620 nm, so that Example Compound No. 1 of the present invention is an optimum compound. On the other hand, Example Compound No. (1-60) of Igarashi is not suitable for this purpose.

(b) The luminescence quantum yield in a toluene solution of Example Compound No. 1 of the present invention is about 2.7 times that of Example Compound No. (1-60) of Igarashi.

(c) A degree of brightness sensed by human eyes varies depending on wavelength of light and is represented by a spectral luminous efficacy. This spectral luminous efficacy is at a maximum at a wavelength of 555 nm. The spectral luminous efficiency is obtained by dividing each of the spectral luminous efficacies by the maximum spectral luminous efficacy. At a larger value of the spectral luminous efficacy, human eyes recognize that the light at the wavelength is brighter. Accordingly, Example Compound No. 1 of the present invention produces luminescence which is about 3 times more sensitive to human

eyes than that of Example Compound No. (1-60) of Igarashi.

8. Comparative Test B

An electroluminescence(EL) device was prepared and evaluated in the same manner as in Example 27 of the present application except that tris (1-phenylisoquinoline-C^2,N)iridium (III) (Example Compound No. 1 of invention) was changed to tris [1-(2-naphtyl)-isoquinoline- C^3,N]iridium (III) (Example Compound No. (1-60) of Igarashi). The results are shown in Table 2 below.

Table 2

	Ex. Comp. No. 1 of the Invention	Ex. Comp. No. (1-60) of Igarashi
(d) CIE(X,Y)	(0.68,0.32)	(0.71,0.27)
(e) EL emission efficiency (cd/A)	5.2	1.5
(f) EL external quantum efficiency (100cd/m^2)	10.5%	3.4%

(d) CIE coordinates for pure red of the three primary colors (red, green, and blue) of the NTSC color system used in organic EL display are CIE(X,Y)=(0.67,0.33), so that Example Compound No. 1 of the present invention provides emission color suitable for red. However, CIE coordinates of Example Compound No. (1-60) of Igarashi largely deviated from those for pure red, so that the emission color of Example Compound No. (1-60) of Igarashi is not used as red.

(e) The EL emission efficiency, represented by luminance per unit current (cd/A), of Example Compound No. 1 of the present invention is about 3.5 times that of Example

Compound No. (1-60) of Igarashi.

(f) The EL external quantum efficiency at 100cd/m^2 of Example Compound No. 1 of the present invention is about 3 times that of Example Compound No. (1-60) of Igarashi.

9. As shown by these test results, unlike Example Compound No. (1-60) of Igarashi, Example Compound No. 1 of the present invention is suitable as a red luminescent material of the practical EL display and unexpectedly provides better luminescence quantum yield and better spectral luminous efficiency. Further, the resultant EL display also unexpectedly provides better EL performances.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Subscribed this 14th day of November, 2005

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